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VERIFICATION OF A TRANSLATION

I, Charles Edward SITCH BA,

Deputy Managing Director of RWS Group Ltd UK Translation Division, of Europa House, Marsham Way, Gerrards Cross, Buckinghamshire, England declare:

That the translator responsible for the attached translation is knowledgeable in the German language in which the below identified international application was filed, and that, to the best of RWS Group Ltd knowledge and belief, the English translation of the international application No. PCT/DE2004/001930 is a true and complete translation of the above identified international application as filed.

I hereby declare that all the statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the patent application issued thereon.

Date: February 8, 2006

Signature :



For and on behalf of RWS Group Ltd

Post Office Address :

Europa House, Marsham Way,  
Gerrards Cross, Buckinghamshire,  
England.

Polymer mixtures for printed polymer electronic circuits

5       Plastics (polymers) are known as insulators. However, there are also some remarkable polymers having  
conductive and even semiconductive properties. All  
three properties taken together permit the production  
of fully functioning integrated circuits from polymers.  
The attraction of polymer electronics lies in its  
10       simple producability because the polymers can be  
deposited from the solution to give layers. This means  
that in particular it is possible to use inexpensive  
printing techniques by means of which the individual  
structured layers of integrated circuits can be  
15       produced. However, every printing process sets  
specific requirements with regard to the substances to  
be printed, in this case the polymer solutions. It is  
rare that the properties of the polymer solutions  
comply with the printing requirements from the outset.  
20       Thus, for example, the viscosity of the polymer  
solutions is considerably too low for most printing  
processes. This applies in particular to polymeric  
semiconductor material.

25       An inkjet technique for printing low-viscosity  
semiconductor material is known from Nalwa H. S.  
(editor): "Organic Conductive Molecules and Polymers",  
volume 2, 1997, pages 334 to 335). Inkjet printing is,  
however, least favored for mass production.

30       Starting from this, it is the object of the invention  
to make semiconductive polymers accessible to standard  
printing processes.

35       This object is achieved by the inventions stated in the  
independent claims. Advantageous developments are  
evident from the dependent claims.

Accordingly, a polymer mixture, in particular a polymer

solution, contains one or more semiconductive polymers and one or more non-semiconductive, i.e. insulating and/or conductive polymers.

5 Polythiophene, in particular poly(3-hexylthiophene) (P3HT), has proven particularly advantageous as a semiconductive polymer. However, the use of polyfluorene or polythienylenevinylene and a mixture of two or three of said semiconductive polymers is also  
10 possible.

Polystyrene (PS), polymethyl methacrylate (PMMA), cymel and polyisobutyl (PIB) or mixtures thereof have proven particularly suitable as non-semiconductive polymers.

15 In order to obtain a polymer solution, solvents may also be present in the polymer mixture, in particular chloroform, toluene, ketones, dioxane and/or heptane.

20 In addition, the polymer mixture may contain conductive polymers, oligomers, conductive molecules and/or semiconductive molecules (monomers, "small molecules", in particular pentacene and/or C60), particles and other materials which cannot be dissolved, or may  
25 consist of a selection of said substances and possibly customary additives.

By said mixing of semiconductive and non-semiconductive polymers, it is possible to establish the desired  
30 viscosity of the polymer solution. Preferably, a viscosity of more than 8 mPa.s is established, in particular more than 80 mPa.s. As a result, the polymer solution is suitable for screen printing or pad printing and further standard printing processes.

35 A polymer mixture of the type described can preferably be used in a printing process, in particular in a screen printing, flexographic printing, offset printing, gravure printing and/or pad printing process.

By means of a polymer mixture of the type described, it is possible to produce a double layer which contains one or more semiconductive polymers in its first layer  
5 and one or more non-semiconductive polymers in its second layer.

This can be effected, for example, in a process for the production of the double layer in which a polymer  
10 mixture of the type described is used, which polymer mixture separates on deposition from the solution and thus forms the double layer.

A printed electronic circuit can be produced using a  
15 polymer mixture of the type described, it being possible to create semiconductive polymer structures by screen printing and/or pad printing during the production. Alternatively or in addition, the polymer mixture can also be used generally for the production  
20 of electrical components, for example for organic transistors, diodes, capacitors, resistors, light emitting diodes, photovoltaic cells, photodetectors, display elements, etc.

25 Preferred developments of the printing process, of the double layer, of the process for the production of the double layer and of the electronic circuit arise from the preferred developments of the polymer mixture, and vice versa.

30 Further advantages and features of the invention are evident from the description of working examples with reference to the drawing.

35 Figure 1 shows the characteristic of an organic field effect transistor having a semiconductive layer which consists purely of semiconductive polymer;

Figure 2 shows the characteristic of an organic field effect transistor having a semiconductive layer which consists of non-semiconductive polymer and semiconductive polymer in the weight ratio 1:3;

Figure 3 shows the characteristic of an organic field effect transistor having a semiconductive layer which consists of non-semiconductive and semiconductive polymer in the weight ratio 1:1.

It is proposed to mix the functional, i.e. semiconductive polymers with other polymers in order in this manner to achieve an adaptation to a wide range of printing requirements. The mixing of polymers (polymer blends) is a customary method for obtaining certain combinations of properties. However, in the case of the semiconductor polymers, such mixtures have not yet been considered. If such a mixture were to be considered theoretically, virtually complete disappearance of the semiconductive properties would be expected for semiconductor polymers as a result of addition of other materials. However, our own experiments show that this is not the case.

Polythiophene, as a semiconductive polymer, was mixed with polystyrene and polymethyl methacrylate. It is found that the semiconductor function is retained in the mixed polymer system too. At the same time, the corresponding polymer solution has an increased viscosity, which makes it more readily printable.

The reason why the semiconductive properties are so well retained is not exactly known, but two arguments appear possible. Firstly, polyaniline is a conductive polymer. Like polythiophene, it belongs to the class of the conjugated polymers. In polymer mixtures, it retains its conductive function to a high degree, as

described in Speakman S. P. et al.: "Organic Electronics 2 (2)", 2001, pages 65 to 73. What is true here for polyaniline may apparently also be applied to polythiophene. Secondly, it is a known phenomenon that  
5 polymer mixtures tend to separate after deposition from solution. This is described, for example, in Garbassi F. et al.: "Polymer Surfaces", 1998, pages 289 to 300. The polymer system minimizes its total energy by virtue of the fact that the material having the lower surface  
10 energy forms the uppermost layer.

In our case, two layers form, the first of which substantially comprises the admixed polymer (PS or PMMA). The second layer is a virtually pure  
15 polythiophene layer. It imparts the semiconductor property to the system. What has been demonstrated here for polythiophene by way of example is also possible with other semiconductive polymers, for example polyfluorene and polythienylenevinylene.

20 Figures 1 to 3 show the characteristics of three organic field effect transistors (OFETs) having different mass ratios of polystyrene (PS) and poly(3-hexylthiophene) (P3HT) in the semiconductive layer. In  
25 figure 1, the ratio of PS to P3HT is 0:100, in figure 2 the ratio of PS to P3HT is 25:75 and in figure 3 the ratio of PS to P3HT is 50:50. The layer thicknesses are unchanged for better comparison.

30 The OFETs comprising PS and P3HT, whose characteristics are shown in figures 2 and 3, function just as well as the OFET comprising P3HT, whose characteristics are shown in figure 1, except that the current decreases with increasing proportion of PS. Since, however, the  
35 OFF current decreases more sharply than the ON current, the transistor characteristic of the ON/OFF ratio even improves.

A further tested working example is the system P3HT and

polyisobutyl (PIB). At maximum solubility of 2.5% of polymer solid in chloroform, P3HT has a viscosity of 2 mPa.s. By adding PIB dissolved in heptane the viscosity can be increased to values up to 100 mPa.s, depending on the mixing ratio. This meets the requirements of screen printing, in which the viscosity must be greater than or equal to 10 mPa.s, and of pad printing, in which the viscosity must be greater than or equal to 100 mPa.s. In experiments, operable OFETs have also been produced with this semiconductor mixture.

By means of the invention, the properties of different polymers are combined with one another. For example, polythiophene contributes the semiconductive property and polystyrene the higher viscosity in the polymer solution. Polythiophene alone in solution would not be readily printable owing to the excessively low viscosity, whereas the addition of polystyrene imparts higher viscosity to the solution, which makes it more readily printable. Further advantages are to be seen for the case of the abovementioned separation. They relate to the solid double layer, for example of insulator and semiconductor, remaining behind after evaporation of the solvent. Specifically, the advantages are as follows:

- saving of an operation by simultaneous production of the two layers,
- production of an extremely thin semiconductor layer, which is not possible by direct printing of only the semiconductor solution,
- perfect bonding (adhesion) of the two layers to one another,
- the layers rest on one another in exact register, particularly in printed structures (self-alignment),
- the solvent compatibility does not play a role, i.e. there is no partial dissolution of the lower

layer on application of the upper layer,  
- it is to be expected that the special method of  
layer formation by separation has a positive  
influence on the layer quality, for example with  
5 regard to low defect density and high structural  
order of the (conjugated) polymers.